

New Environmentally-Friendly Materials

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REVIEW

Curated Topics on Novel Exotic Eco-Friendly Materials for Sustainable Rechargeable Battery Technologies

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ABSTRACT

This work explores novel and eco-friendly materials that hold transformative potential in addressing the limitations of traditional rechargeable battery systems while aligning with sustainability goals. Beginning with a brief overview of global energy demand and rechargeable battery architectures, this curated study delves into four cutting-edge materials classes, each offering unique advantages. Nanocomposites for Silicon-based anodes represent a breakthrough in enhancing energy density and addressing volume expansion issues, which are the major limitations of silicon anodes. We explore the possibility of recycling silicon from photovoltaic (PV) panels and its associated Life Cycle Assessment (LCA) costs. The study then shifts focus to the realm of two-dimensional (2D) materials, highlighting their exceptional electrical, mechanical, and catalytic properties, which promise substantial improvements in (Li, Na, K-ion based) battery performance. Metal-organic frameworks (MOFs), with their tunable porosity and multifunctionality, are investigated for their role in polysulfide confinement and catalytic enhancement in lithium-sulfur and other battery chemistries. The next topic delves

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Copyright © 2024 by the author(s). Published by Japan Bilingual Publishing Co. This is an open access article under the Creative Commons Attribution 4.0 International (CC BY 4.0) License (https://creativecommons.org/licenses/by/4.0/). into bio-based catalysts, which have emerged as sustainable alternatives for facilitating electrochemical reactions, leveraging renewable resources to minimize environmental impact. Finally, conductive polymers are briefly considered for their ability to offer flexibility and conductivity, possibility of getting rid of inactive materials in batteries, paving the way for advanced, deformable energy storage devices. This compilation underscores the immense potential of these exotic materials in revolutionizing battery technologies, providing insights into their applications, challenges, and scalability. The discussion concludes with future perspectives on integrating these materials into commercial systems to achieve energy sustainability. *Keywords:* Global Energy Demand; Sustainable Practices; Novel Materials; Silicon-Based Anodes; Conductive Polymers; Metal-Organic Frameworks; Two-Dimensional Materials; Nanocomposites

1. Introduction

1.1. Benchmarking Global Energy Demand

In a quest to benchmark the technical capabilities of potential extraterrestrial civilizations, Kardashev^[1, 2] proposed a new scale based on energy consumption, which is named after him. According to this Kardashev scale, Type-1 refers to the civilizations capable of harnessing all the energy sources of a planet, while Type 2 and 3 refers to those civilizations, whose energy demands are of the level of the native star and galaxy respectively. In this Kardashev scale, humans are still at the level of 'Civilization Type 1.0'.

Presently our primary energy sources include fossil fuels, renewables, and fissile materials available on earth. Transitioning from unsustainable fossil fuels to renewable energy sources is significantly increasing the demand for critical minerals. They are needed for technologies like solar panels and EV batteries, wind turbines etc. The International Energy Agency (IEA) predicts a six-fold rise in mineral demand by 2050, highlighting the strong link between economic growth and energy demand^[3]. Three defining societal challenges that presently dominate the scientific discourses are: climate change, energy security, and economic stability.

Globalization, economic growth, and technological progress have increased energy demands dramatically, necessitating a re-evaluation of the distribution of energy, food, and water for sustainable development^[4]. The global energy sector is undergoing a profound transformation fueled by technological innovations as well as, unfortunately with geopolitical tensions. These shifts are reshaping production and consumption patterns, trade flows, and the roles of energy exporters and importers^[5]. Traditionally, countries are categorized as energy exporters or importers based on their energy trade balance, with exporters exhibiting a negative balance and importers a positive one. A more pragmatic approach will be to consider it holistically at a global scale, as the world is progressively transitioning from nation-centric systems to a more network-centric approach of global institutions and organizations. Therefore, it becomes increasingly important to comprehend these shifts to effectively adapt to the evolving economic and political dynamics^[6].

1.2. A Quick Primer on the Internal Architecture of Batteries and Curated Topics

Rechargeable batteries consist of a well-organized internal structure designed to facilitate the controlled movement of ions and electrons to store and release energy efficiently. Below is an overview of their internal architecture (see Figure 1). An electrochemical cell consists of two electrical terminals: an anode and a cathode, together they are called electrodes. These electrodes are separated by an electrolyte and a thin barrier called a separator. The separator allows the electrolyte to pass through but keeps the electrodes from touching each other [7, 8]. The energy density of a battery is expressed in two main ways: gravimetric energy density and volumetric energy density. Gravimetric energy density, also known as specific energy, measures the energy content relative to the battery's weight and is expressed in Watt-hours per kilogram (W-hr/kg). Volumetric energy density measures the energy content relative to the battery's volume and is expressed in Watt-hours per liter (W-hr/l). It is directly proportional to the product of the net electrochemical potential difference between the electrodes and the battery's charge storage capacity. Batteries with electrodes capable of high charge storage and electrolytes with a broad electrochemical stability window exhibit higher energy densities, making them ideal for advanced energy storage applications^[9].



Figure 1. Schematic description of rechargeable batteries. (a) The working principle of the rechargeable battery (b) Representation of the C/LiCoO₂ cell. Reproduced from reference^[10, 11].

In this article, we are curating a few topics that have a high potential to revolutionize rechargeable batteries. The first topic focuses on nanocomposite silicon anodes for Li-ion batteries. Given the limited and uneven global distribution of lithium salts, challenges related to cost and potential supply chain disruptions are expected in the near future^[12]. Silicon is highly regarded as a potential anode material for lithium ion batteries (LIBs) due to its theoretical capacity, which greatly exceeds that of graphite, the prevalent material in current commercial LIBs^[13, 14]. However, the transition to silicon anodes in LIBs faces several challenges, primarily due to the significant volume expansion during lithiation and delithiation. This expansion leads to severe mechanical strain, crack formation, and the eventual loss of active material, resulting in poor cycle life and performance degradation. To address these issues, nanocomposite silicon anodes have emerged as a promising solution^[15, 16].

This article explores two particularly innovative approaches to nanocomposite silicon anodes. The critical question is whether these materials can be sourced sustainably through recycling or not. Recycling silicon, especially from end-of-life photovoltaic (PV) panels and other waste streams is essential to reduce carbon footprints, promoting environmental sustainability, and emphasizing the importance of life cycle assessment (LCA) in material processing. Despite the environmental benefits, significant economic challenges such as high operational costs and limited scalability, remain barriers to the widespread adoption of recycling processes under current conditions^[17].

Another intriguing approach involves bio-derived silicon composites, particularly from agricultural waste like rice husks and bamboo leaves. These materials offer a costeffective and sustainable pathway for silicon anode production, providing a viable short-term solution before advanced bottom-up fabrication methods become economically feasible. Leveraging bio-derived materials not only addresses sustainability concerns but also opens opportunities for reducing waste and developing efficient recycling methodologies^[18, 19] Together, these strategies highlight the immense potential of nanocomposite silicon anodes in shaping the next generation of high-performance, eco-friendly batteries.

However, in the longer run the bottom-up approach is touted to revolutionize the future of manufacturing. The lower dimensional materials have the capacity of embedding functionality in the materials itself. In traditional manufacturing, the functionality of a product was typically achieved by the assembly of parts. Therefore, miniaturization was limited by the physical size limits of the parts. In this new bottomup approach, since properties are embedded in the materials itself, it can be miniaturized nearly to the molecular size, if there is such a requirement. It offers unparalleled design flexibility in product development. Therefore, the low dimensional materials and metal organic frameworks (MOFs) are likely to dominate the future discourse of these materials' development. So, this is chosen as the other curated topic for this article^[20, 21].

Investigating and exploring bio-based materials and catalysts is another important direction in all applied mate-

rials research and rechargeable batteries are no exception. This topic, of course, is another curated focus for this article. The potential of bio-based materials to revolutionize battery technology, by offering sustainable and environmentally friendly alternatives, makes it a compelling area of research and development.

In the pursuit of enhancing overall performance of batteries, getting rid of non-functional (dead) materials while simultaneously improving safety are of utmost importance. Conductive polymers (CPs) fit in this bill very nicely. It can be used to replace the separators, improve the performance of electrodes and many more, and has been chosen as another curated topic.

2. Nanocomposites for Si-Anode

Silicon, an element from group 14 in the periodic table, played a crucial role in the development of the modern world, particularly ushering in the electronics revolution and Industry 4.0. It is the second most abundant element in the earth's crust and usually occurs in oxide form. As a metalloid, silicon exhibits some properties of metals and some of non-metals. At room temperature, silicon has a bandgap of 1.12 eV which indicates it is a semiconductor in nature^[22, 23]. Due to its stable operation as a semiconductor, silicon is widely used in the semiconductor industry to construct a wide range of electrical components including transistors, integrated circuits, solar panels, and sensors^[24-26]. The thermal conductivity of silicon varies with the composition and the method of fabrication. Generally, high thermal conductivity is crucial in the case of high-performance electronic and photonic devices, silicon and its compound with carbon are designed to provide good thermal conductivity at room temperature^[27-29]. Further, silicon is used to make a wide range of alloy materials, such as aluminum-silicon and ferrosilicon. These alloys are used to make dynamos, transformer plates, engine blocks, and machine tools^[30, 31].

In addition to a wide range of applications in various industries, silicon is also being used as an anode material for secondary storage devices^[32]. Since silicon is also an abundant material in the earth's crust, the price of silicon when compared with other alloy-based anode materials is cheaper^[33–35]. These advantages make silicon a much better anode material for secondary storage devices. Currently,

LIBs are dominating the market of secondary storage devices due to their robust, stable, and long cycling capabilities^[36]. Initially, secondary batteries were prepared for small-scale portable electronic devices [37, 38]. However, with the increasing demand for energy and to reduce carbon emissions it is expected that the global battery demand will be 2600 GWh by 2030^[39]. This may cause a surge in the price of LIBs as lithium sources are finite and geographically restricted in limited countries^[40]. Further, recycling LIBs is an energyintensive and costly process, which can cause additional carbon emission^[41]. To reduce carbon footprint due to the extraction and recycling of LIB components for secondary storage devices, it is necessary to find alternative anode materials that can deliver large specific capacities without compromising the cost and performance of the battery. Silicon, an eco-friendly material, which facilitates high specific capacity at low cost particularly in the alloy form of Li-Si, can be a good alternative to lithium metal anode to develop sustainable energy storage devices^[42, 43]. It is to be noted here that, when compared with conventional anode material, which is graphite, silicon delivers specific capacity of nearly ten times higher than that of graphite and it also has low discharge potential^[14, 34].

Despite the advantages, Silicon as an anode faces some issues that are hindering the commercialization of Si-based LIBs. These challenges include high-volume expansion, low intrinsic electronic conductivity, unstable solid electrolyte interphase (SEI) formation, and delamination from electrodes after pulverization^[44-46]. To resolve these issues, several strategies have been proposed by the scientific community. The issue of volume expansion can be mitigated by implementing porous nanostructures, nanoparticles, or nanowires^[34, 47–49]. Further, the intrinsic low conductivity of silicon can be avoided by constructing silicon-carbon composite anode materials^[50]. With the use of additives to the electrolyte, stable SEI can be formed which will provide stability and good coulombic efficiency^[51, 52]. Since the scope of this article is focused on sustainable nanocomposite for Si anode, we will consider only eco-friendly materials that are beneficial to constructing Si-based anode for sustainable energy storage devices. Towards that end, in section 2.1 we will discuss silicon and silicene (a silicon allotrope) composites derived from recycled PVs, which is expected to produce a huge amount of wastages in the near future because of its

rapid adaptation from falling prices (waste in the range of 60–78 million tons by 2050). Section 2.2 will be focused on the Life Cycle Assessment (LCA) to highlight key challenges associated with the current recycling process of silicon from PV cells. At the end of this topic, in section 2.3 we will specifically discuss two promising examples of bio-derived silicon composites harnessed from rice husks and bamboo leaves.

2.1. Recycled Silicon-Based Composites from Photovoltaic (PV) Panels

It is expected that the volume of PV panel waste will be in the range of 60–78 million tons by 2050^[53]. The adoption of PVs is rapidly expanding because of falling prices, so much so, that it is now cheaper to produce electricity through it than fossil fuels in many instances^[54]. With such rapid and explosive growth, the waste production and its recycling will be an acute issue in the future. Therefore, it is imperative to find good use cases in some applications that have similar growth potential. Recycled silicon-based composite anodes are gaining attention as a sustainable and efficient solution for secondary storage devices like LIBs^[55]. These anodes are produced by repurposing silicon extracted from end-of-life electronics or solar panels, which helps reduce electronic waste and supports a circular economy^[56]. By integrating recycled silicon with materials such as carbon, graphene, or various polymers, these composites overcome challenges such as silicon's inherent volume expansion and mechanical breakdown during cycling, thereby improving the overall performance and longevity of the anodes^[57].

Therefore, it is quite necessary to recycle silicon and other materials from PV panel wastage and utilize them for further applications. The utilization of recycled silicon helps to lower the production costs significantly while maintaining a high specific capacity, which greatly exceeds that of traditional graphite anodes.

To demonstrate the effectiveness of the recycled silicon for its potential use as anode for secondary storage devices, several studies have been carried out. Small silicon particles that are generated as the leftovers of the silicon wafer cutting process, one of the byproducts of the PV industry, is known as Kerf loss silicon (KL-Si). Wu et al.^[58] developed a Si-SiC-Ni (SSN) composite material from KL-Si. Among the tested materials, the SSN composite having Si-to-Ni weight

ratio of 3:2 (SSN-3-2) exhibited outstanding cycle stability and a reduced volume expansion of 39%, compared to the 287% observed in the KL electrode. Yin et al.^[59] developed a method using micro sized KL-Si combined with melamine and formaldehyde, to obtain a melamine-formaldehyde (MF) coating on KL-Si in the presence of acetic acid. This composite is then processed through high-temperature pyrolysis, resulting in porous carbon-coated silicon (Si@SiO₂@C), where silicon and carbon layers were separated by a thin SiO₂ layer. To create a hollow Si@void@C structure, the material was etched with 10% HF for 10 minutes, removing specific components to achieve the desired morphology. The obtained anode composite delivered a capacity of 1164.4 mAh g^{-1} after 300 cycles at rate of 1 A g^{-1} . At high current rate of 3 Ag^{-1} , the composite delivered capacity of 927 mAh g^{-1} after 500 cycles. Further, the practical application of this composite was tested by employing NCM622 as a cathode to construct a full cell which retained a capacity of 81.5% after 150 cycles. In a recent study, Yin et al.^[60] utilized ZIF90 as a carbon source and obtained KL-SiC-ZIF/N/Co composite as anode which delivered reversible capacity of 981 mAh g^{-1} and capacity retention of 81.9% at the rate of 2 A g^{-1} after 350 cycles. Tang et al.^[61] used a freeze-drying process to obtain SiC anode from KL-Si. The enhancement in the porosity due to the freeze-drying process provided pathways for Li-diffusion and enhanced performance of the anode material. The obtained anode material delivered a capacity of 792 mAh g^{-1} at rate of 0.1 A g^{-1} after 100 cycles.

Similarly, Kanaphan et al.^[62] extracted multilayer silicene, a silicon allotrope, from discarded solar cells through chemical activation and exfoliation techniques, creating an anode material that maintained a capacity of 290 mAh g⁻¹ over 500 cycles at a rate of 1C. Mahmoud et al.^[63] successfully recovered silicon from discarded PVs by leaching process to eliminate impurities, followed by ball milling to produce nanostructured silicon for use as an anode material. Silicon ball-milled at 250 RPM for 10 hours delivered a remarkable capacity of 1400 mAh g⁻¹ at 0.05C after 50 cycles. Similarly, Liu et al.^[64] recycled silicon from end-of-life solar panels, creating W-pSi@C/CNT composites for anodes. These materials achieved an outstanding capacity of 2040 mAh g⁻¹ at a rate of 0.2 A g⁻¹ after 200 cycles, highlighting their potential in sustainable energy storage. Recycling silicon waste from the PV sector not only minimizes its environmental impact but also contributes to the advancement of sustainable and high-performance energy storage technologies. As a result, recycled silicon-based composite anodes stand out as vital components in the development of ecofriendly technologies and renewable energy solutions.

2.2. Life Cycle Assessment (LCA) of Recovering Silicon from PV Panels

The average lifespan of crystalline PV panels ranges from 25 to 30 years^[65, 66]. When these panels reach the end of their lifecycle, they can be recycled to recover valuable materials, including silicon, aluminum, copper, glass, and silver. However, extracting high-purity materials, especially silicon, is both energy-intensive and technically challenging. While more complex processing techniques can improve silicon quality, they also increase associated costs. Therefore, exploring alternative applications for recycled silicon is essential^[67].

Currently, the irregular and low volume of PV waste makes recycling uneconomical compared to landfill disposal fees^[68]. For potential applications, like using recycled silicon as an anode material in batteries, it is essential to conduct a LCA before establishing large-scale recycling operations. LCA evaluates the environmental impacts of manufacturing or recycling processes, considering factors like energy use, water consumption, greenhouse gas emissions, and waste generation^[69]. Incorporating LCA into silicon recycling provides valuable insights into designing sustainable processes.

Studies like those by Latunussa et al.^[69] have shown that recycling silicon PV panels through advanced physical and chemical treatments offers significant environmental benefits compared to traditional disposal methods. Their findings suggest that as solar panel installations continue to grow, recycling PV waste will become increasingly important. Similarly, Dias et al.^[70] conducted LCA and life cycle cost analysis (LCCA) on silicon extraction using organic solvent delamination, finding the processes to be environmentally friendly but not yet cost-effective.

The cost challenges of recycling compared to landfill disposal are anticipated to diminish with the large-scale deployment of solar panels, which will generate higher volumes of PV waste. Moreover, ongoing advancements in cost-effective recycling methods are expected to further reduce costs, enhancing the economic feasibility of recycling processes. More government policy support towards waste utilization is desirable.

2.3. Bio-Derived Silicon Composites: Rice Husk and Bamboo Leaves

The extraction of silicon from agricultural waste or from natural resources is an efficient way to minimize carbon footprint when compared with extraction from mines primarily because it will require excessive nano structuring or other approaches to cope with repeated volume expansioncontraction cycles. One of the key sources of silicon from agricultural waste is rice husk. Globally, annual rice husk production is around 150 million tons which is considered agriculture waste^[71]. After burning this rice husk in air, the yield of silica is around 20% of the dry weight of rice husk^[72]. This ash of rice husk, with 85–95% of silica, is highly porous, lightweight, and possesses a large surface area^[73, 74]. The extraction of silica from rice husk can be carried out by different methods which involves thermal treatment, hydrothermal treatment and chemical treatment^[75]. On obtaining silica, pure form of silicon can be obtained by following magnesium thermal, Zincothermic reduction reaction^[76–78].

As silicon anodes go through extreme volume expansion during charging and discharging, utilization of the naturally porous structure of rice husk to obtain porous anode material is an effective solution. By utilizing such a porous structure, nano size porous silicon nanoparticles (SiNP's) having size of 10-40 nm was obtained and incorporated as anode material in LIB. The obtained anode delivers a high specific capacity of 1750 mAh g^{-1} at rate of C/2^[80]. In another approach, after obtaining micro-size porous SiO₂/C composite from rice husk, ZnCl₂ was used to activate rice husk ash to obtain a high surface area of $1191.30 \text{ m}^2\text{g}^{-1}$. The increased surface area provided extra contact between active materials and electrolyte which increased Li-ion diffusion pathways to deliver an excellent reversible capacity of 1105 mAh g^{-1} after 360 cycles at current density of 0.1 A g^{-1} ^[81]. Zhang et al.^[82] utilized rice husk as precursor to obtain SiNP's having size of 50 nm which then compose with N-doped carbon and CNT (SNCC) in order obtain 3D hierarchical hybrid structure of composite spheres. These spheres provided a high reversible capacity of 1380 mAh g^{-1} at 0.5 A g⁻¹. Autthawong et al.^[79] utilized rice husk as source of silica and synthesized ternary components based composite

material by comprising tin (Sn) and bronze-titanium dioxide $TiO_2(B)$ with SiO_2 . As shown in the HRTEM images of **Figure 2**, the uniform distribution of Sn-SiO₂ on the surface of $TiO_2(B)$ nanorods enabled fast charge transfer without

causing excess volume expansion. The obtained composite material provided excellent cycle life of 500 cycles at current density of 5 A g^{-1} and delivered specific capacity of 143.03 mAh g^{-1} without any failure.



Figure 2. Electrochemical performance of as-prepared TiO₂(B), SiO₂@TiO₂(B), and Sn–SiO₂@TiO₂(B). (**a**) rate cycle capability at different current densities in the range of 50–5000 mA g⁻¹; (**b**) long-term cycle stability and the corresponding Coulombic efficiency at a fast-charging state of 5000 mA g⁻¹ for 500 cycles; (**c**) CV curves of first-three cycles of the Sn–SiO₂@TiO₂ electrode between 0.01 and 3.0 V at a scan rate of 0.2 mV s⁻¹; (**d**) Nyquist plots of as-prepared electrodes; (**e**) HR-TEM images with the lattice fringes of Sn–SiO₂@TiO₂(B) and the low-magnification TEM image inset. Reproduced from reference^[79].

In addition to rice husks, bamboo leaves also contain high amounts of silicon and possess 3D porous structure. Considering this fact, Yu et al.^[83] constructed 3D porous Si/N-doped carbon composite as anode material for LIBs which delivered maximum capacity of 360 mAh g^{-1} after 120 cycles at current density of 1.6 A g^{-1} . Pang et al.^[84] constructed a ternary Ni/SiOx/nitrogen-doped carbon (NSC) composite from bamboo leaves by in-situ transformation of a Ni₃Si₂O₅(OH)₄/nitrogen-doped carbon. Implementation of uniformly distributed Ni nanoparticles on SiOx/nitrogendoped carbon framework reduced volume expansion and improved conductivity of the electrode. The obtained material delivered a capacity of 864.6 mAh g^{-1} at 0.2A g^{-1} after 70 cycles and exhibited superior rate capability of 289.8 mAh g^{-1} at 10 A g^{-1} . The high capacity and superior performance delivered by bio-derived Si-based composite assures the quality and reliability towards its use as anode for energy storage devices.

3. Low Dimensional (2D and MOFs) Materials

The emergence of two-dimensional (2D) materials represents a groundbreaking advancement in materials science, particularly in the field of sustainable rechargeable battery technologies^[34, 85, 86]. These materials, characterized by their single or few-layer structures, exhibit unique properties that can enhance energy storage capabilities. Among the most studied 2D materials are graphene, Carbon nanotubes (CNTs) transition metal dichalcogenides (TMDs), MOFs, and MXenes. Their distinct physical and chemical attributes allow for significant improvements in battery performance, addressing the urgent need for eco-friendly energy solutions^[87]. This discussion explores how these exotic materials can revolutionize battery technology while promoting environmental sustainability.

3.1. 2D Materials

Graphene, a monolayer of carbon atoms arranged in a hexagonal lattice, is the most well-known 2D material. Its exceptional electrical conductivity, mechanical strength, and thermal properties make it an attractive candidate for battery applications^[88, 89]. Research has shown that incorporating graphene into LIBs can significantly enhance their charge capacity and cycling stability^[90]. However, the challenge remains in producing high-quality graphene at scale. Ecofriendly synthesis methods, such as liquid-phase exfoliation and chemical vapor deposition, are being investigated to minimize environmental impacts. The integration of graphene into battery electrodes not only improves performance but also paves the way for the use of more sustainable materials in battery construction.

CNTs have garnered significant attention in battery technology due to their unique structural, electrical, and mechanical properties. Their high electrical conductivity and large specific surface area make them excellent conductive additives in Li-ion and next-generation batteries^[91]. CNTs improve electron transport within electrodes, reducing internal resistance and enhancing rate capability^[92]. Additionally, their ability to form a robust conductive network supports the integration of high-capacity active materials, such as silicon or sulfur, which often suffer from poor conductivity and significant volume changes during cycling. For instance, CNTs have been employed as hosts in sulfur cathodes for lithiumsulfur (Li-S) batteries to trap polysulfides and mitigate the shuttle effect, thus improving cycling stability^[92]. Moreover, CNTs contribute to the mechanical stability of electrodes. Their flexibility and tensile strength buffer against the stresses induced by material expansion and contraction, particularly in silicon anodes and other alloy-type materials. They have also been incorporated into current collectors and separators, enhancing the overall structural integrity and thermal stability of the battery. Beyond the traditional Li-ion systems, CNTs have demonstrated potential in solid-state and flexible batteries, offering pathways for integration into next-generation energy storage devices^[93]. Their compatibility with advanced electrolytes and architectures makes them versatile candidates for cutting-edge battery designs^[94].

TMDs, including materials like molybdenum disulfide (MoS₂) and tungsten diselenide (WSe₂), have emerged as

strong contenders in the quest for sustainable battery technologies^[95]. TMDs exhibit tunable band gaps and high surface areas, allowing for efficient ion intercalation and enhanced charge/discharge rates^[96]. Studies demonstrate that MoS₂ can improve energy density and cycling stability when used in LIB anodes^[97, 98]. The ability to modify the electronic properties of TMDs through chemical doping and structural alterations provides opportunities for creating customized battery systems tailored to specific performance needs^[99]. This adaptability, coupled with environmentally friendly synthesis methods, positions TMDs as an asset in sustainable battery technology.

3.2. 3D Printing of Batteries/Components Using 2D Materials

In recent years, both industry and academics have shown a great deal of interest in the additive manufacturing (AM) process, also referred to as three-dimensional (3D) printing. For the development of future low-cost, high throughput printed electronics, graphene and other 2D materials are crucial. Advances in ink formulations for screen, gravure, inkjet, and extrusion printing have boosted interest in 2D material-based inks, enabling efficient production across various substrates and extrusion-based printing excels in creating diverse 3D structures^[95, 96]. According to Rojaee et al.^[100] 2D materials enhance ion flux through interfaces, block lithium polysulphide species, prevent electrode pulverization, improve Li+ ion deposition, boost thermal stability, and aid in the formation and decomposition of LiO₂ discharge products. This promotes the development of safe lithium batteries with high energy and power density. The integration of 2D materials into 3D printed battery designs has emerged as a promising avenue for enhancing the performance and scalability of energy storage devices. 2D materials like graphene, MoS₂, and other transition metal dichalcogenides (TMDs) possess exceptional electrical conductivity, high surface area, and mechanical properties, making them ideal candidates for next-generation batteries. When these materials are combined with 3D printing technology, they allow for the creation of highly efficient, customizable, and complex battery architectures. This synergy not only improves the electrochemical performance but also allows for the development of batteries with enhanced energy density, rapid charge/discharge capabilities, and longer cycle life. 3.3. Metal Organic Frameworks (MOFs) The use of 2D materials in 3D printed batteries is transforming the landscape of energy storage solutions. 3D printing offers unparalleled precision in the design of battery electrodes and other components, enabling the fabrication of intricate geometries that are impossible to achieve with traditional manufacturing methods. This flexibility in design is a key advantage in creating batteries that can be tailored for specific applications, such as flexible electronics or electric vehicles (EVs). For instance, the incorporation of graphene and TMDs into 3D printed structures has demonstrated significant improvements in the structural integrity and electrochemical stability of batteries. 3D printing allows for the development of hierarchical structures that maximize the interaction between the 2D materials and the electrolyte, which leads to improved charge/discharge kinetics and cycle stability^[101].

Another exciting aspect of 3D printed batteries using 2D materials is their potential for scalability. Traditional manufacturing techniques for batteries often face limitations in producing large-scale, high-performance devices. However, 3D printing allows to produce batteries with high precision at lower costs, making them more accessible for commercial use. 3D printing combined with 2D materials can lead to batteries with a higher volumetric energy density, which is crucial for applications in energy storage systems where space and weight are at a premium. Additionally, the possibility of printing batteries in flexible and stretchable forms further enhances the applicability of these devices in wearable electronics and other portable devices^[100].

The research into 3D printed batteries using 2D materials is still evolving, with new findings and improvements emerging regularly. Ongoing work is focused on optimizing the interactions between the 2D materials and the other components of the battery, such as the electrolyte and current collectors. Research highlighted in [102] emphasizes the importance of fine-tuning the printing processes and the materials used to achieve a balance between electrical conductivity, mechanical stability, and electrochemical performance. As these technologies mature, it is expected that 3D printed batteries incorporating 2D materials will play a critical role in the development of sustainable and high-performance energy storage solutions, paving the way for more efficient and cost-effective energy systems.

MOFs are a class of advanced materials formed from the coordination of metal ions with organic ligands, creating a highly porous structure^[86]. These materials exhibit remarkable properties, such as an exceptionally high surface area that can exceed 6000 m^2/g , making them attractive for a variety of applications. The tunability of their pore sizes and structures allows for selective adsorption and storage of gases, separation of mixtures, and even catalysis, and energy storage applications positioning MOFs as versatile tools in materials science and engineering^[103, 104]. MOFs have emerged as highly promising materials for battery applications, due to their unique structural characteristics and tunable properties. Their large surface area, high porosity, and the ability to host a variety of metal centers make them excellent candidates for improving the performance of energy storage devices like lithium-ion, sodium-ion, potassium ion, and even solid-state batteries^[105]. The crystalline nature of some of the MOFs allows them to accommodate a variety of electroactive species and facilitate the efficient transport of ions, which is crucial for enhancing battery capacity, charge/discharge rates, and overall stability. The ability to modify the organic linkers and metal sites in MOFs offers further opportunities to tailor their electronic and ionic conductivity, making them adaptable to different types of battery chemistries^[106]. In LIBs, for instance, MOFs can serve as either cathode, anode, or electrolyte additive materials^[107]. When used as anode materials, MOFs can offer high capacity due to the incorporation of metal ions that can undergo reversible redox reactions^[34]. The porosity of MOFs also plays a role in accommodating the volume changes that occur during cycling, helping to preserve the structural integrity of the anode^[76]. Additionally, certain MOFs can be combined with carbon materials to enhance their conductivity, a key requirement for improving the rate performance of batteries^[108]. The structural versatility of MOFs also allows for the incorporation of additional functionalities, such as hosting electrolyte additives or stabilizing other electrochemical components, which can further enhance battery efficiency.

In sodium-ion battery (SIB) and other post-lithium battery systems, MOFs offer an attractive alternative due to their ability to host larger ions, such as sodium, in their pores. This enables them to accommodate the unique requirements of these battery chemistries, which often face challenges

related to the size and mobility of the ions involved ^[109]. The flexible and adaptable nature of MOFs allows for the design of materials that can optimize ion diffusion rates and enhance cycle stability, which are critical factors for next-generation energy storage technologies. Moreover, their potential for incorporating other redox-active species, such as transition metal clusters, offers additional avenues for improving the overall energy density and cycling life of batteries ^[110].

However, despite the promising outlook, several challenges need to be addressed for MOFs to reach their full potential in battery applications. Issues such as the inherent instability of some MOFs under certain electrochemical conditions, as well as difficulties in scaling up their synthesis for large-scale battery production, remain significant barriers^[111]. To overcome these challenges, researchers are exploring ways to enhance the structural stability of MOFs through surface modifications, hybridization with other materials, and the development of more robust synthetic routes. With ongoing advancements, MOFs hold great promise for revolutionizing the energy storage sector, offering efficient, sustainable, and high-performance alternatives to traditional battery materials^[112].

MOFs have demonstrated immense potential in addressing the key challenges faced by Li-S batteries, such as the polysulfide shuttle effect, poor conductivity, and limited cycle life^[92, 113]. MOFs act as sulfur hosts or catalytic materials. leveraging their high porosity, large surface area, and tunable chemical functionalities to enhance battery performance^[114]. For example, ZIF-8, a zinc-based MOF, has been employed as a sulfur host due to its ability to physically confine polysulfides and prevent their diffusion to the anode^[113]. Similarly, MOFs such as MIL-101(Cr), a chromium-based MOF, when functionalized with CPs, significantly enhances sulfur utilization in Li-S batteries. The functionalization enables efficient redox reactions during charge/discharge processes by improving electronic conductivity and facilitating ion diffusion. This synergy addresses one of the critical challenges in Li-S batteries polysulfide shuttling thereby improving overall battery efficiency and cycle life. These findings underscore the potential of MOF-based materials in enhancing electrochemical performance^[115]. Another notable example is MOF-derived carbon frameworks like HKUST-1, named after the place of discovery: Hong Kong University of Sci-

ence and Technology, year 1999 by Chui et al.^[116] Upon pyrolysis, HKUST-1 undergoes structural transformation to yield a highly conductive carbon matrix capable of accommodating sulfur. This carbon matrix not only enhances sulfur loading but also preserves the structural integrity of the electrode during repeated cycling. The improved conductivity and structural stability directly contribute to better sulfur utilization and capacity retention, addressing key limitations of traditional sulfur cathodes^[117].

In addition to sulfur hosting, MOFs doped with catalytically active metals (e.g., cobalt, copper, or nickel) enhance the conversion kinetics of lithium polysulfides, reducing energy losses. For instance, Co-MOF composites integrated into the cathode architecture have shown improved capacity retention over long cycles^[118]. Furthermore, MOF-based hybrid materials, such as MOF-derived metal oxides and carbon frameworks, have garnered significant attention due to their dual benefits of polysulfide trapping and catalytic activity. MOF-derived materials leverage their parent MOFs' structural precision and chemical diversity, enabling the design of advanced hosts that tackle multiple challenges in Li-S batteries^[119].

For instance, MOF-derived metal oxides, such as cobalt oxide (Co₃O₄) or manganese dioxide (MnO₂) derived from Co-MOF and Mn-MOF precursors, have shown excellent capability in trapping lithium polysulfides through chemical interactions while catalyzing redox conversion. Co₃O₄ nanoparticles, when uniformly dispersed in carbon matrices derived from MOFs, effectively suppress the polysulfide shuttle and enhance reaction kinetics, as demonstrated in studies where the batteries exhibited extended cycling stability and high sulfur utilization^[120, 121]. Similarly, MnO₂-based hybrid materials, derived from MIL-100(Mn), provide abundant active sites for the immobilization of polysulfides and boost their catalytic conversion, resulting in improved discharge capacities and minimized capacity fade over extended cycles^[122]. The advancements underline the versatility of MOFs in Li-S batteries, demonstrating their ability to combine favorable properties like high capacity, improved cycle life, and environmental sustainability, making them a cornerstone of next-generation energy storage technologies. Figure 3 shows the MOFs applications for different batteries.



Figure 3. MOF coated Si and Sb alloy type anode materials for Li and post Li-ion battery applications.

4. Bio-Based Materials and Catalysts

The rapid growth of EVs has driven advances in LIBs, with Li-S batteries gaining attention for their high energy density and low cost. However, challenges like sulfur cathode cycling and the shuttle effect hinder their progress. Biomassderived materials offer a sustainable and efficient solution due to their natural abundance, low cost, and superior adsorption capabilities, making them ideal for developing innovative bio-carbons with hierarchical porosity^[123]. Bioaerogels, derived from renewable sources like cellulose and lignin, offer high surface area, mesoporosity, and biodegradability, making them sustainable alternatives for energy storage. Their use as electrodes and separators enhances energy density, power density, and cycle life, with recent advancements highlighting their potential as anode materials for LIBs^[124]. Spinel CoFe₂O₄-decorated bio-derived N-doped carbon enhances oxygen electrocatalysis through strong metal-substrate interaction, enabling lithium-air batteries with high discharge capacity (18356 mAhg⁻¹) and stability over 200 cycles^[125]. All-solid-state batteries with solvent-free polymeric electrolytes promise high energy density and safety, but improving room-temperature ionic conductivity and interfacial charge transport remains crucial. Embedding mineral and biobased fillers, derived from natural resources, enhances solvent-free polymeric electrolyte performance while promoting sustainability, paving the way for greener energy solutions (see **Figure 4**)^[126].

Biomass-based separators, especially cellulose-based membranes, offer excellent electrolyte wettability, mechanical strength, and stability, with the added benefits of biodegradability and abundant functional groups, making them promising for battery applications. However, challenges remain in optimizing their performance for commercial use^[127]. Research shows that surface coating significantly improves the thermal stability and electrolyte wettability of polyolefin separators^[130]. For example, Kim et al.^[128, 129] developed a trilayer separator with polydopamine and graphene-carboxymethyl cellulose coatings, enhancing electrolyte wettability and electrochemical stability. Similarly, calcium alginate fibers with intrinsic flame retardancy are fabricated using wet spinning of sodium alginate, offering further advancements in separator design^[131]. Thermal stability is vital for separators to prevent short circuits, with boron nitride nanotubes (BNNTs) emerging as a highperformance solution for improved safety^[132].



Figure 4. Prospects of applications of bio-based materials in rechargeable batteries. (a) Bio-based and mineral additives for cutting-edge solid polymer electrolytes, and (b) benefits, limitations, mechanisms, and compositions of mineral and bio-based fillers for enhancing the electrochemical performance of solid polymer electrolytes (SPEs). Reproduced from reference^[126].

Biomass-derived carbon materials, synthesized from sources like cotton, silk, wood, and fibers, offer sustainable and cost-effective alternatives to petroleum-based precursors for metal-air battery electrodes. Their high surface area, functional groups, and 3D mesh structure provide excellent conductivity, flexibility, and electrochemical performance, making them ideal for next-generation flexible energy storage devices^[133, 134]. Biomass-derived carbon materials, including nitrogen-doped catalysts produced via pyrolysis, acid treatment, and heteroatom doping, enhance LIB performance by offering high electrocatalytic activity and faster reaction kinetics^[135–137].

Expanding research on biomass-based separators is essential to overcoming current challenges and unlocking their full potential for commercial battery applications, paving the way for sustainable and eco-friendly energy storage solutions. Bio-based binder systems, such as lignin, sodium alginate, cellulose, and carboxymethyl cellulose (CMC), have shown promise in lithium-ion battery cathodes like LiFePO₄(LFP). These systems demonstrated comparable performance to PVDF, with capacities around 140 mAh g^{-1} at 0.1 C (the 'C' stands for the current rate adjusted in such a way that the battery is charged/discharged within an hour), while modifications, such as pH adjustments and binder combinations, improved stability and cycle life^[138]. Bio-based binders like lignin and cellulose show promise in LIBs, offering eco-friendly alternatives to polyvinylidene fluoride (PVDF). Advances in stability and conductivity have enabled over 1000 cycles, but challenges in durability, scalability, and cost remain as issues of concern for commercial use.

5. Conductive Polymers (CPs)

The increasing depletion of fossil fuels, escalating energy costs, and growing environmental concerns have underscored the urgency of reducing carbon emissions and adopting sustainable energy storage technologies, including supercapacitors, batteries, and solar cells. CPs have emerged as promising materials due to their high electron affinity, redox activity, ease of processing, corrosion resistance, cost-effectiveness, and excellent electrical conductivity^[139]. These polymers have a backbone structure with alternating single and double bonds, allowing for delocalized π -electrons that confer unique optical and electronic properties, as illustrated in **Figure 5**.

The field of CPs gained momentum with Alan G. Mac-Diarmid's discovery of enhanced conductivity in sulfur nitride (SN)x upon bromine doping, a breakthrough that significantly advanced the study of conducting materials. This pioneering work also demonstrated that doping polyacetylene with bromine could enhance its conductivity by several orders of magnitude, ultimately earning the Nobel prize in chemistry in 2000^[140]. PVDF, with its high fluorine content, necessitates the use of toxic and costly NMP solvents for electrode processing and requires high drying temperatures, making the process environmentally unfriendly. Alternatives like low-fluorine poly(ionic liquids) or anionic single-ion lithium-conducting polymers offer improved cycling stability and capacity in Li-air and solid-state batteries^[141, 142]. Among CPs, materials such as polypyrrole (PPy), polyaniline (PANI), polythiophene (PTh), poly (3,4stand out for their remarkable conductivity and stability, mak-

ethylenedioxythiophene) (PEDOT), and their derivatives ing them strong candidates for energy storage applications in batteries^[143].





Non-polyether-based SPEs offer advantages over traditional polyether systems, particularly in terms of electrochemical stability, compatibility with high-voltage electrodes, and performance at elevated temperatures. Polyether-based SPEs (e.g., PEO) have limitations like poor ionic conductivity at room temperature, reduced stability at high voltages, and limited compatibility with high-performance electrodes. To overcome these challenges, alternative polymers such as carbonyl-containing polymers, ionomer-based polymers (e.g., poly (ionic liquids)), fluorine-based, and nitrile-based polymers are being explored for improved ionic conductivity, stability, and electrode compatibility in LIBs^[144].

CPs have shown potential as cathode materials for LIBs, but their low doping levels and poor cycling stability limit their effectiveness. A novel approach using nanoporeconfined in situ electropolymerization to create nanostructured polythiophene-type porous cathodes has addressed these issues to some extent by improving doping availability and cycling stability. The resulting thieno[3,2-b]thiophene (TtTP)/active carbon cathode demonstrates an ultrahigh reversible capacity of 309.2 mAh g^{-1} , an energy density of 1252.3 Wh kg⁻¹, and exceptional rate capability, outperforming other conducting polymers and p-type organic cathodes^[145]. Figure 6 showcases the CPs based on their conductivity.



Figure 6. CPs classified based on their conductivity mechanisms. Adapted from Reference^[146].

In Li-S batteries, the polysulfide shuttle effect reduces active material utilization and causes capacity fading. Conductive polymer-based interlayers between the cathode and separator mitigate this issue by anchoring, confining, and converting soluble polysulfides, improving battery performance. The interlayer mass loading and the electrochemical window (1.7–2.8 V, sometimes 1.5–3 V) play crucial roles in determining the capacity and cycling stability of Li-S batteries^[147].

The successful application of CPs relies on efficient material handling, polymerization, and enhancing electrode properties, with recent advances focusing on hybrid composites for improved performance^[146].

6. Author's Perspective

The future evolution of the energy storage system will be propelled by transformative advances in materials science, manufacturing techniques, and system integration. As the global shift toward renewable energy accelerates, the demand for more efficient, sustainable, and cost-effective energy storage solutions will become increasingly urgent. The integration of emerging technologies, including artificial intelligence, additive manufacturing, and bio-based materials, will significantly enhance the performance, sustainability, and recyclability of energy storage systems. These innovations will improve energy density, cycle life, and overall efficiency, while also addressing environmental concerns related to material sourcing and waste. Additionally, advances in system integration will enable seamless interactions between storage devices, power grids, and renewable energy sources, paving the way for more resilient and flexible energy infrastructures.

In the coming years, a collaborative approach involving industry, academia, and policymakers will be crucial in overcoming the technical and economic challenges facing energy storage systems. By fostering interdisciplinary partnerships, driving innovation, and aligning regulatory frameworks, we can ensure a smooth transition to a low-carbon, energy-efficient future, where energy storage systems play a critical role in powering sustainable societies and industries worldwide.

The advancement of silicon-based composite anodes presents promising opportunities for sustainable energy stor-

age. Future research should prioritize enhancing the scalability and efficiency of silicon extraction from renewable and recycled sources, such as agricultural by-products and discarded electronic devices. Innovative approaches, such as developing hybrid composites and nanoengineered designs, can effectively address challenges like volume expansion and low conductivity, enabling the production of long-lasting and efficient batteries. MOFs and 2D materials in battery applications are highly promising, with the potential to overcome critical limitations in current energy storage technologies. Future research will focus on scalable production methods, environmental sustainability, and innovative designs that leverage their properties to develop high-performance, costeffective, and durable battery technologies for EVs, portable electronics, and renewable energy storage systems.

Additionally, incorporating eco-friendly processes and materials, such as biodegradable binders and bio-derived carbon, can significantly improve the environmental sustainability of these systems. A coordinated effort among industries, researchers, and policymakers will be critical to achieving large-scale implementation and aligning these developments with global sustainability initiatives.

7. Conclusions

This review underscores the transformative potential of novel and eco-friendly materials in advancing rechargeable battery technologies to meet global energy demands sustainably. Through a curated exploration of cutting-edge materials, including nanocomposites for silicon anodes, 2D materials, MOFs, and conductive polymers, the study highlights how these innovations address key challenges such as capacity limitations, environmental impact, and mechanical degradation. Each material class demonstrates unique advantages, from enhanced energy density and conductivity to improved cyclic stability and scalability, offering a glimpse into the future of sustainable energy storage systems.

Global research efforts in energy storage systems are focused on enhancing key factors such as high storage capacity, lightweight design, cost-effectiveness, long-term stability, and design flexibility. With the increasing global demand for energy and the drive towards sustainability, advancements in rechargeable battery technologies, including Li-ion and Li-S batteries, are essential for enabling the transition to renewable energy sources. Innovations in battery components, such as conducting polymers, bio-based catalysts, and advanced separators, are improving energy density, cycle life, and sustainability, while also reducing environmental footprints. As the energy landscape shifts, a combination of technological innovation, strategic policies, and education is needed to promote energy efficiency and minimize environmental impacts. The integration of energy-efficient devices like supercapacitors, alongside the use of artificial intelligence and additive manufacturing, will further optimize energy storage and reduce environmental harm. Ultimately, enhancing battery efficiency, sustainability, and recycling will be crucial for supporting a greener energy future and addressing the challenges of climate change, energy security, and economic stability.

Silicon-based composite anodes created from biosourced and recycled materials represent a significant advancement in LIB technology. Their combination of high specific capacity, cost efficiency, and environmental sustainability positions them as strong contenders to replace traditional graphite-based anodes. Through advanced material engineering, challenges such as volume expansion and structural degradation can be largely addressed, leading to improved battery durability and performance. Incorporating bio-derived and recycled silicon supports global objectives to reduce waste and lower carbon emissions, contributing to the shift toward greener energy storage solutions. As key components of next-generation batteries, silicon-based composites are poised to drive progress in renewable energy and sustainable technology development. Bio-based materials provide a sustainable, eco-friendly alternative for diverse applications, combining renewability and biodegradability to support a circular economy. Advances in research and innovations have enhanced their performance and scalability, making them competitive with traditional materials. Progressive works in research are essential to achieving their full potential and tackling global issues like climate change and resource depletion. Conducting polymers show significant potential as cathode materials for lithium-ion batteries, but their practical application has been limited by issues like low doping levels and poor cycling stability. The introduction of nanostructured polythiophene-type porous cathodes

through nanopore-confined in situ electropolymerization offers a promising approach, enhancing doping efficiency and cycling stability, thereby improving the feasibility of these materials for future energy storage technologies.

While the promising properties of these materials are evident, realizing their commercial viability requires overcoming challenges related to cost, scalability, and integration into existing manufacturing processes. Ultimately, this review emphasizes the critical role of innovative materials in reshaping battery technologies to align with the dual objectives of energy sustainability and technological advancement, paving the way for a greener, energy-efficient future.

Author Contributions

Conceptualization, A.K., A.N., S.R., H.S., and K.K.S.; writing—original draft preparation A.K., A.N., S.R., H.S., and K.K.S.; writing—review and editing A.K., A.N., S.R., H.S., and K.K.S.; supervision K.K.S. All authors have read and agreed to the published version of the manuscript.

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